

wherein the condensed phase comprises at least one polycyclic aromatic hydrocarbon having from ten to thirty carbon atoms. - -

REMARKS

Claim Amendments

Claims 1, 46 and 49 have been amended to more clearly define the invention. The amendments to claims 1, 46 and 49 have a basis at page 15, line 1 of the specification.

Rejection under 35 USC §112

Claims 1-28, 34-42, 44 and 46-52 have been rejected under 35 U.S.C. § 112, first paragraph, as containing subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the art that the inventors has possession of the claimed invention at the time the application was filed. The Office Action states that there is no disclosure of pyrene being formed in the liquid phase in a flame as it is a combustible hydrocarbon and that page 13 of the specification casts doubt on whether all hydrocarbon species form liquid phases in a flame.

It is respectfully submitted that pyrene and the other polycyclic aromatic hydrocarbon species recited in the claims do indeed form liquid phases in a flame. The evidence for this exists in Figures 3, 5 and 6 of the application as originally filed.

As detailed at page 25 of the specification, two thousand particles collected from a flame were subjected to analysis by mass spectroscopy. Then 1440 spectra were

divided into six bins of 240 spectra each. The spectra in each bin were averaged. The results are presented in Figure 5 of the application.

In Figure 5, the results from the lowest ion count bin shown in panel (a) depict very sharp well-defined polycyclic aromatic hydrocarbon (PAH) and hydrogenated PAH lines at  $m/z$  190, 202, 204, 216, 228, 230, 240, 242, 254, 264, 266, 276, 278, 290, and 302. (Pyrene is within this  $m/z$  range.) This spectrum represents essentially 240 "uncarbonized" PAH-containing particles. As the RIC increases, the amount of carbonaceous soot slowly increases over the first four panels, (a-d). The intensities of the masses immediately surrounding the PAH peaks dramatically increase over the same range. This is evidence that the hydrogen atoms on the PAHs are becoming very labile and rapidly exchanging between PAHs, leaving them susceptible to rearrangement. This is evidence of pyrolytic addition of small hydrocarbons to the existing PAHs in the particle. In panels (e) and (f), the PAH intensities decrease whereas the intensities of the carbon cluster peaks (see left hand side of panel) increase at the same rate. Therefore, the correlation between the rate of PAH ion loss and the rate of carbon cluster ion gain is more than a mere trend. This suggests that the carbonaceous or mature soot is being created directly from the PAHs in the particle.

The averaged mass spectra of the "uncarbonized" PAH- containing particles (Fig. 5a) are reproduced in Fig. 6 of the application on a larger scale to permit a detailed analysis of its component ions. The possible chemical formula for each of the major ions is shown above its respective mass. The high-mass side of the spectrum is populated by PAH ions, and the low-mass side is populated by oxygenated fragment ions, hydrocarbon fragment ions, and carbon cluster ions. Close inspection of the high-

mass side of the spectrum yields insight into the composition and chemistry of the PAH-containing particles present in the flame because of the nature of the ions present as well as the nature of the ions that are not.

Because alkylated PAHs are not stable in the condensed phase in the flame, the most abundant PAH ion in the "uncarbonized" PAH-containing particle spectrum ( $m/z$  216,  $C_{17}H_{14}$ ) has a five-membered ring. This suggests that, under the conditions in the flame where the PAH-containing particles are being formed and growing, pentagonal ring structures are thermodynamically stable. Moreover, examination of the carbonization evolution spectra in Fig. 5 suggests that the five-membered ring species survive in the condensed phase up to the point where carbonization occurs.

One of the most striking aspects of the "uncarbonized" PAH-containing particle spectrum is the lack of ion intensity at  $m/z$  252. This mass represents the benzo[a]pyrene class of PAHs. However, there is a corresponding peak at  $m/z$  254. Logically, the only possible explanation is that the benzo[a]pyrene class of PAHs is hydrogenated. Perusal of the PAH ion distribution in Fig. 6 shows the presence of other hydrogenated species at  $m/z$  204, 230, 242, 266, 278, 302, and 304. The peaks at  $m/z$  254 and 302 are much more intense than their parent PAH ions at  $m/z$  252 and 300 as is typically seen in the spectra of ethene soot. These PAHs are preferentially hydrogenated. This phenomenon can not be explained by gas-phase chemistry because the rate of hydrogenation, in that case, is dependent on the number of aromatic rings. Therefore, the degree of hydrogenation would increase with increasing PAH mass if the hydrogenation process were the result of gas-phase chemistry. Consequently, the observed preferential hydrogenation must be the result of flame-

induced chemistry in the condensed phase. Because the PAHs in the measured distribution from the PAH-containing particles show signs of hydrogenation and this hydrogenation is the result of condensed-phase chemistry, the PAH-containing particles must have an abundance of labile hydrogen to produce hydrogenated PAHs.

Because of the local Gaussian distribution around each of the PAH masses, it is evident that where one PAH loses a hydrogen atom another PAH gains it. Where a hydrogen atom is lost, a radical is formed. Where a hydrogen atom is gained, a radical is formed. This means that there is a substantial concentration of radicals in the PAH-containing particles when they are being pyrolyzed by the flame. It is evident that these radicals are being formed with a very low activation energy - much lower than currently thought possible by those people who base their models on gas-phase reaction kinetics.

Another consequence of rapid hydrogen exchange in the PAH-containing particles is the ability of that medium to act as a hydrogen sink. Because the PAHs can rapidly exchange hydrogen, they can also store it for a time when it is added by an outside source. The process of carbon-based mass growth involves the loss of two hydrogen atoms and therefore can act as an outside source of hydrogen. This is the reason for the excess labile hydrogen that results in the preferential hydrogenation of the PAHs that was observed in the "uncarbonized" PAH-containing particle mass spectra (see Figure 6).

Normalized aerodynamic size distribution histograms show that PAH-containing and mature soot particles have the same size distributions. This means that the rates of condensation and desorption of gas-phase species are the same for all the particles across the entire cross section of the flame. Because the diffusion flame temperature

increases in going from the inside to the outside of the flame, it also suggests that these same rates are essentially temperature-independent. There is only one way for this observation to be correct - the condensation step must be irreversible over the measured temperature range. The correctness of this observation is supported by the measured PAH distribution from the soot particles. The boiling points (i.e., the temperature where the vapor pressure is equal to the atmospheric pressure) of the most populous PAHs are well exceeded when these same PAHs are condensing to form the PAH-containing droplets. This means that the condensation process must involve the formation of chemical bonds. In other words, chemical condensation is occurring. Van der Waals interactions are not sufficient under these conditions. The temperature-independent irreversibility of the condensation process explains the reason for the relatively narrow size distribution seen in Figure 3 of the application. This temperature independence is the result of the low activation energy for creation of radicals in the soot particle phase.

In summary, the physical evidence presented above and in Figures 3, 5, and 6 of the application, by itself, is conclusive proof that these micron-sized, optically transparent, PAH-containing particles exist in the flame even above the boiling point of the hydrocarbon species when analyzed. Thus, it is respectfully requested that the rejection under 35 USC §112 be withdrawn.

Rejection under 35 USC §102(e) or 35 USC §103(a)

Claims 1-28, 34-42, 44 and 46-52 were rejected under 35 USC §102(e) or 35 USC §103(a) over U.S. Patent No. 5,985,232 to Howard et al. ("Howard"). The Office

Action states that Howard teaches forming a combustion flame and collecting polyatomic [sic polyaromatic] hydrocarbon condensibles from a liquid.

Amended independent claims 1, 46 and 49 now recite that the carbon-containing material is condensed in the flame. This amendment serves to more clearly distinguish the claimed invention from Howard.

In the process of Howard, an unsaturated hydrocarbon fuel and oxygen are combusted in a non-arc-discharge burner chamber to establish a flame, and condensibles of the flame are collected at a post-flame location. Throughout Howard, it is disclosed that the condensibles of the flame are collected at a post-flame location. (See page 2, lines 26-28; page 2, lines 41-42; page 3, lines 5-8; page 3, lines 19-21; and page 3, lines 24-26 of Howard). Furthermore, the process by which Howard collects condensibles of the flame is described at column 7, lines 41-54 wherein it is stated:

Samples of soot and condensibles were collected at a given "post-flame" distance from the burner. Post-flame collection sites included two and seven centimeters "down stream" from the burner, as measured perpendicular from the burner surface to the orifice at the tip of the sampling probe. Vapor collection and quenching was accomplished by inserting a water-cooled quartz probe connected to a filter, vacuum pump, and gas meter into the flame vapor stream, rapidly withdrawing and cooling the vapors and collecting the condensate ("condensibles") on the in-line filter. Sample of condensibles were also collected from the water-cooled top surface of the combustion chamber against which the flame tail impinged.

Thus, the condensibles of the Howard process are being collected by way of the physical condensation of the flame vapor stream, typically by use of a cooled probe.

In contrast, the invention recited in claims 1, 46 and 49 includes the step of collecting at least a portion of a condensed phase comprising a liquid phase in the

flame. Thus, there is a significant difference between the present invention and the Howard process. Specifically, the Howard process is relying on physical condensation of a vapor to collect condensibles. In the present invention, chemical condensation in the flame forms a condensed phase (see discussion above) and this condensed phase is thereafter collected from the flame. Accordingly, claims 1, 46 and 49 have been amended to make it clear that the carbon-containing material is condensed in the flame before collection of the condensed phase from the flame.

Because of the differences in the collection processes of the present invention and Howard, different materials are collected in the processes. By condensing the flame vapor stream at a downstream location, the Howard process will collect mature soot, fullerenes, nanotubes and gas phase condensed hydrocarbons. In contrast, the method of the present invention collects from a flame a condensed phase comprising at least one polycyclic aromatic hydrocarbon that may be used to form fullerenes and nanotubes in subsequent steps.

In addition, the precursor soot produced in Howard's flame is converted into mature soot before it exits the flame. The fullerenes that Howard sees likely derive from the gas phase and during the carbonization of precursor soot. The fullerenes produced in Howard's flame represent only a couple of percent of the product. Most of the product is mature black soot. In contrast, the present invention serves to thwart the production of mature soot and collect primarily precursor soot and use that as a reactive medium to produce fullerenes by subsequent manipulation after the flame. Thus, further differences in Howard's process and the present invention are evident.

Because Howard fails to teach or suggest all of the limitations of claims 1, 46 and 49, it is believed that claims 1, 46 and 49 (and claims 2-28, 34-42, 44, 47-48 and 50-52 that depend thereon) are patentable over U.S. Patent No. 5,985,232 to Howard et al.

Conclusion

It is respectfully submitted that the entire application has been placed in condition for allowance. Favorable reconsideration is respectfully requested. No fees are believed to be needed for this amendment. However, if fees are needed, please charge them to Deposit Account 17-0055.

Respectfully submitted,  
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Dated: February 10, 2003

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**Version with markings to show changes made**

-- 1. (Three Times Amended) A method for producing a polycyclic aromatic hydrocarbon comprising:

a. combusting a carbon-containing material in a flame to condense the carbon-containing material in the flame; and

b. collecting at least a portion of a condensed phase comprising a liquid phase in the flame;

wherein the condensed phase comprises at least one polycyclic aromatic hydrocarbon. --

-- 46. (Amended) A method for producing a polycyclic aromatic hydrocarbon, the method comprising:

a. combusting at least one carbon-containing material in a flame to condense the carbon-containing material in the flame; and

b. collecting at least a portion of a condensed phase comprising a liquid phase in the flame;

wherein the condensed phase comprises pyrene. --

- - 49. (Amended) A method for producing a polycyclic aromatic hydrocarbon, the method comprising:

a. combusting a carbon-containing material in a flame to condense the carbon-containing material in the flame; and

b. collecting at least a portion of a condensed phase comprising a liquid phase in the flame;

wherein the condensed phase comprises at least one polycyclic aromatic hydrocarbon having from ten to thirty carbon atoms. - -